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Determination of Optical Birefringence of S-Benzylisothiouronium Complex Crystals using Channelled Spectrum Method

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Abstract

The channeled spectrum method was used for measuring the birefringence of an anisotropic material. Crystals of uniform thickness were placed between crossed polarizers and illuminated with a tungsten source. The resulting pattern was analyzed to deduce the birefringence and its dispersion across the visible spectrum. Cauchy's two parameter formula was applied to fit the experimental data. The Cauchy's coefficients A and B were obtained by solving the linear equations using MATLAB. The value of birefringence was also calculated.

Keywords : *Anisotropic materials; MATLAB; Birefringence.*

1.INTRODUCTION

The ability of some organic crystals to surpass the performance of current inorganic materials in optical devices prompts much of the research into the design and the synthesis of new organic crystals that possess powerful nonlinear optical properties. Large non-linearities have been observed in organic materials containing unsaturated molecules, where the valence electrons of some of the atoms are most completely paired in localized bonds, often the molecule is aromatic, containing benzene or similar rings and it is the theory of the electronic structure of such systems that provides the basis for analysis of their non-linear properties. The structural characteristic of S-benzylisothiouronium chloride(SBTC)and S-benzylisothiouronium nitrate (SBTN), on the molecular scale, reveals that they are π-conjugated and possess good planar structure. The electrons in the π-system are more mobile than those in the σ-frame work and are therefore more easily polarisable. Since the πelectrons are less tightly bound, the spacing of their energy levels and the transition energies are smaller than for σ electrons. The complete delocalization of πelectrons of the ring systems makes them wholly aromatic in character. Due to the longer π-electron conjugation, the compounds exhibit high birefringence.

An isotropic optical material has only one refractive index but an anisotropic material may have two or three refractive indices. With two refractive indices, the material behaves optically as a uniaxial crystal, but for three refractive indices the material behaves optically as a biaxial crystal. The difference between higher and lower refractive indices is taken as the birefringence "Δn" of the material. Various methods were used for measuring the birefringence. Methods such as interference

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colours and Berek Compensator (Bloss, 1961), Moire deflection technique (Heller et al. 1985), double speckle photography (Bernar do et al. 1987) and standard compensating method (Chuanzeng et al. 1990) are used for measuring the birefringence. However most of these techniques require expensive and highly sophisticated optical apparatus as well as a large volume sample (Zheng et al. 1992). Channeled spectra (Born et al. 1983) resulting from two-beam interference were used for measuring the anomalous dispersion of various optical materials such as quartz and sapphire.

The crystals used in this research were grown by the slow evaporation method at room temperature. SBTC and SBTN crystallize in orthorhombic space group Pbca and monoclinic space group $P2_1/C$ respectively. The most extensively studied properties of these new materials are the second-order nonlinear optical properties that include frequency doubling and the linear electro-optic effect. Although both effects arise from quadratic electric susceptibility, the optimum arrangement of the molecule in the crystal lattice is different for each effect. In the case of molecular organic crystals the molecule produces the effect predominantly along one axis called polar axis. These crystals have been identified as frequency doublers.

2. CAUCHY'S THEORY

A plane polarized beam incident in the direction of the normal to the anisotropic material is divided into two beams propagating with different velocities. One with vibration parallel to the surface of the optical axis and the other with vibration perpendicular to it. After emergence from the sample, the two beams are recombined again and obtain by the analyzer. Interference fringes are viewed through the telescope. Assuming that the refractive indices of the sample for the two beams are ordinary "n_o" and extraordinary "n_e", and its thickness is "t", then the phase difference between the two beams is given by :

$$\varphi = \left(\frac{2\pi}{\lambda}\right) t(n_o - n_e) \qquad (1)$$

and the condition for a dark fringe is [9]:

$$t\,(n_o - n_e) = n\,\lambda \qquad (2)$$

where "n" is the order of the interference fringes and "λ" is the wavelength of the light used. It is necessary that the sample has uniform thickness "t" and $(n_o - n_e)$ over the whole spectrograph slit. After "P" fringes in the direction of shorter wavelength equation (2) can be written as:

$$\Delta n\, t = (P+n)\,\lambda \qquad (3)$$

where Än is the birefringence. The Cauchy dispersion formula for an anisotropic optical material can be written as

$$\Delta n = A + \frac{B}{\lambda^2} \qquad (4)$$

where A and B are constants characterizing the optical material. This equation represents the curves in the visible region, with considerable accuracy. To find the values of the two constants, it is necessary to know the values of Δn for two different λ's.

3.EXPERIMENTAL METHOD

Anisotropic crystals have crystallography distinct axes and interact with light by a mechanism that is dependent upon the orientation of the crystalline lattice with respect to incident angle. When the light enters the optical axis of anisotropic crystal, it behaves in a manner similar to the interaction with isotropic crystal, and passes through a non equivalent axis; it is refracted into two rays polarized with the vibration directions oriented at right angles to one another, and traveling at different velocities. This phenomenon is termed as double refraction or birefringence and is exhibited to a greater or lesser degree in all anisotropic crystals.

The birefringence of the crystals was measured using channel spectrum method with a high power tungsten lamp (500W) as a source. The

polarizer and analyzer were placed in crossed positions and the crystal was placed, with its optics axis perpendicular to the incident ray. The transmitted light components from the analyzer interfere and an interference pattern was observed through the high resolution spectrometer. For each band, the corresponding minimum deviation angle was noted then the wavelength of corresponding deviated dark band was obtained from the calibration graph of standard wavelength (mercury) spectrum. The birefringence was calculated as

$$\Delta n = \left(\frac{\lambda_2}{(\lambda_1 - \lambda_2)} \right) * \left(\frac{\lambda_1}{t} \right) \qquad (5)$$

where, Δn is the birefringence, t is the thickness of the crystal, λ_1 and λ_2 are the wavelengths.

3. RESULTS AND DISCUSSION

The high resolution spectrometer used in this study was calibrated for minimum deviation angle of the necessary spectrum. Samples are thinned for required size. The thickness of the S-benzylisothiouronium chloride crystal (0.28mm)S-benzylisothiouronium nitrate crystal (0.21mm) was used for measuring birefringence. The sample was placed between the crossed polarizers. The crystal was rotated till its optic axis was perpendicular to the incident ray. The dark and bright fringes were obtained in the visible region. Since the fringes are due to two-beam interference, the widths of the bright and dark fringes are equal. The resulting two-beam interference fringes are serially numbered from zero to n. Then the corresponding wavelength for each fringe is deduced from the calibration graph. On applying the Cauchy's two term model to the experimental data; a set of equations were obtained as given below. The equations were solved using MATLAB 7 software and the coefficients A and B values determined.

$$t\left[A + \frac{B}{\lambda_1^{\,2}} \right] = n_1 \lambda_1 \qquad (6)$$

$$t\left[A + \frac{B}{\lambda_2^2} \right] = n_2 + P\lambda_2 \qquad (7)$$

$$t\left[A + \frac{B}{\lambda_3^2} \right] = n_3 + 2P\lambda_3 \qquad (8)$$

where "t" is the thickness of the sample, n_1, n_2 and n_3 is the fringe order corresponding to the wavelengths λ_1, λ_2 and λ_3. Finally, substituting these values in equation 4, "Δn" is found for different wavelengths across the visible spectrum.

The parameters of a Cauchy dispersion function for the investigated samples are shown in Table 1. Fig.1. shows the wavelength Vs fringe order obtained for SBTC. Fig 2. represents the variation of "Δn" with the wavelength in the range from 5500 to 6725 ú across the visible spectrum applying Cauchy function. For the wavelengths 5500 and 6725 ú the corresponding birefringence values are 0.347 and 0.346 respectively. There is slight decrease in the birefringence values with increase in wavelength. The wavelength Vs fringe order obtained for SBTN is shown in the Fig 3. The variation of "Δn" with the wavelength in the range from 5425 to 6800 ú across the visible spectrum applying Cauchy function is represented in Fig.4. The birefringence values of SBTN for the wavelengths 5425 and 6800 ú are 0.386 and 0.330 respectively. There is significant change in the birefringence values. It is obvious that, for the two samples the birefringence "Δn" is inversely proportional to the wavelength "λ". Using linear fit the correlation coefficient was found to be 0.9997 and 0.9972 for SBTC and SBTN respectively. There is so significant change between the experimental wavelength and the linear fit values. So the calculation was carried out for the experimental wavelength.

Table 1. Parameters of a Cauchy dispersion function

Material	A	$B \times 10^{-14}$
SBTC	0.3445	0.96
SBTN	0.2307	1.4596

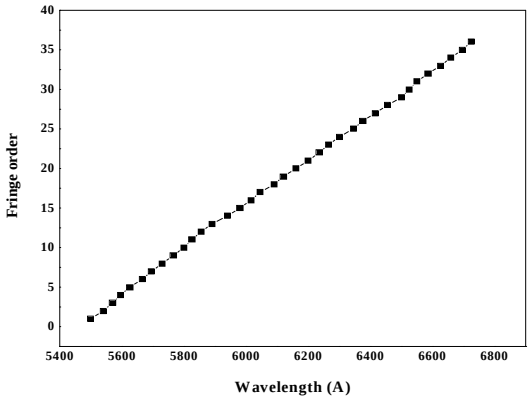

Fig 1. Wavelength Vs Fringe order for SBTC

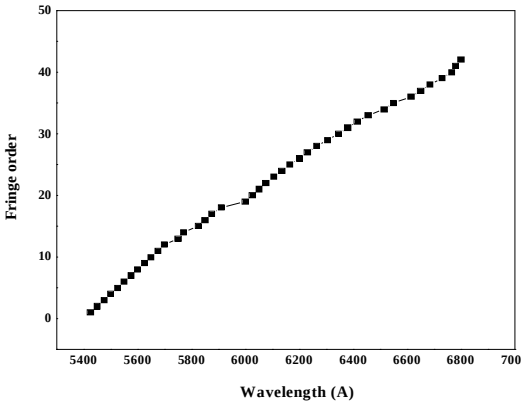

Fig 3. Wavelength Vs Fringe order for SBTN

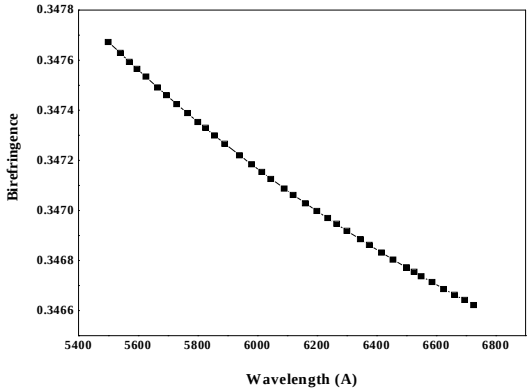

Fig 2. Wavelength Vs birefringence for SBTC

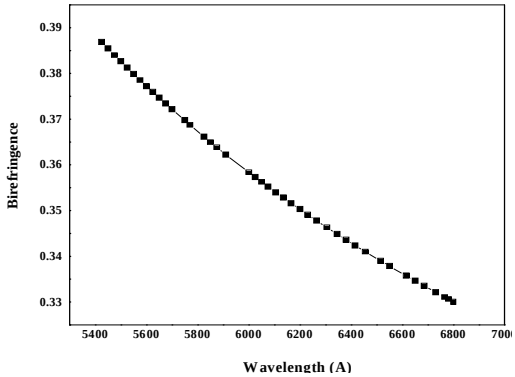

Fig 4. Wavelength Vs Birefringence for SBTN

CONCLUSION

In this work, Channeled spectrum method was used for measuring the natural birefringence and its dispersion across the visible region of spectrum. A two term Cauchy model was used todetermine absolute fringe order. The birefringence value for SBTN is greater than SBTC. These two samples possess negative birefringence.

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